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(54) Title: ORGANIC ELECTROLUMINESCENCE DEVICE AND ANTHRACENE  
DERIVATIVE

(57) Abstract: An organic electroluminescence device comprising a negative electrode and a positive electrode and, interposed therebetween, one or two or more organic thin-film layers including at least a luminescent layer, wherein at least one of the organic thin-film layers contains an anthracene derivative of specified structure added alone or as a component of mixture; and an anthracene derivative of asymmetric specified structure. There are provided an organic electroluminescence device of high luminescence efficiency and long life and an anthracene derivative for realizing the same.

### Technological field

The present invention concerns organic electroluminescent devices and anthracene derivatives, and more specifically concerns organic electroluminescent devices having a high luminescence efficiency and long life, as well as anthracene derivatives making such devices possible.

#### Background technology

The organic electroluminescent devices (hereafter, electroluminescence is abbreviated EL) are self-illuminating devices utilizing the principle of emission of light from phosphors by recombination energy between positive holes injected by a positive electrode and electrons injected by a negative electrode upon the application of an electric field. Since the report of C.W. Tang et al.: Low-voltage organic EL devices by laminated devices (C.W. Tang, S.A. Vanslyke: Applied Physics Letters, vol 51, 913, 1987) of the Eastman Kodak Co., many studies have been carried out concerning organic EL devices using organic substances as constituting materials. Tang et al. used tris(8-hydroxyquinolyl)aluminum in the luminescence layer and a triphenyldiamine derivative in the positive-hole transport layer. Advantages of a laminated structure include an enhanced injection efficiency of the positive holes into the luminescence layer, enhanced efficiency of the formation of excitons formed by recombination, blocking the electrons injected from the negative electrode, capture of excitons formed in the luminescence layer, etc. As shown in this example, a two-layer type of positive hole transport (injection) layer and electron transport luminescence layer, three-layer type of positive hole transport (injection) layer, luminescence layer, and electron transport (injection) layer, etc., are well known for the device structure of organic EL devices. In such laminate-type-structure devices, the device structure and formation processes have been studied for enhancing the recombination efficiency of the injected positive holes and electrons.

Also, chelate complexes such as tris(8-quinolinolato)aluminum complex, etc., coumarin derivatives, tetraphenylbutadiene derivatives, bisstyrylarylene derivatives, oxadiazole derivatives, etc., have been known as luminescent materials. It has been reported that visible light ranging from blue to red can be obtained from them, and the realization of color display devices is expected (e.g., Japanese Kokai Patent No. HEI 8[1996]-239655, HEI 7[1995]-138561, HEI 3[1991]-200289, etc.).

Also, devices using phenylanthracene derivatives as luminescent materials have been disclosed in Japanese Kokai Patent No. HEI 8[1996]-012600. Such anthracene derivatives are used as blue [-light] emitting materials, while a longer device service life is desirable. Furthermore, device materials having fluoranthene groups at the 9 and 10 positions of anthracene were disclosed in Japanese Kokai Patent No. 2001-257074. These anthracene derivatives are also blue emitting materials, yet an improved device service life is necessary. Use of various

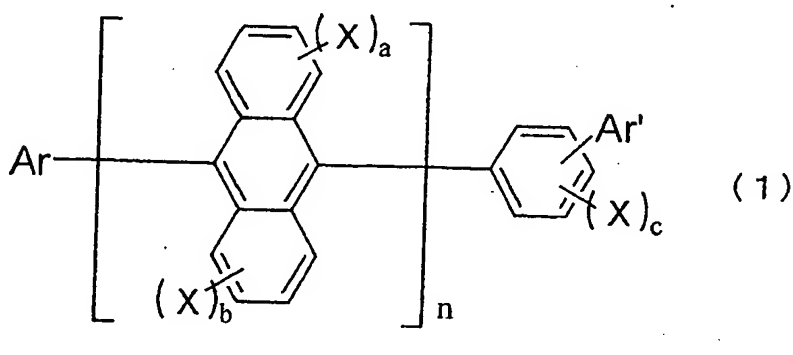
anthracene derivatives for positive-hole transporting materials was disclosed in Japanese Kokai Patent No. 2000-182776. However, their syntheses have not been carried out in practice, and their evaluation as luminescent materials is incomplete.

#### Disclosure of the invention

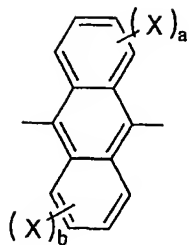
It is an objective of the present invention to overcome such problems and to provide anthracene derivatives enabling the realization of long-life organic EL devices with a high luminescence efficiency.

As a result of an intense study for achieving such objectives, we have discovered that long-life organic EL devices with a high luminescence efficiency can be obtained by using anthracene compounds having a certain asymmetric structure shown by general formula (1) or (2) as luminescence materials of organic EL devices. Thus, the present invention is attained.

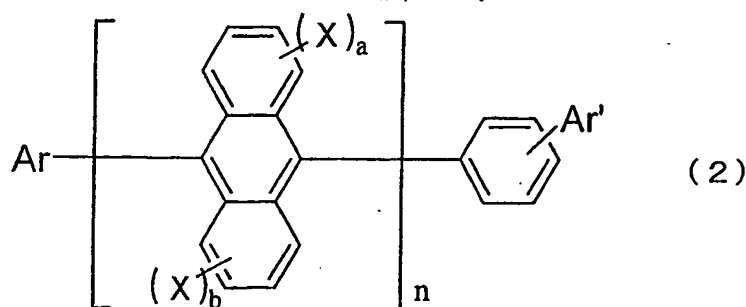
Namely, the present invention provides organic electroluminescence devices having at least one luminescence-layer-containing organic thin film layer sandwiched between a positive electrode and negative electrode, wherein at least one layer of the organic thin film layer contains the anthracene derivative represented by general formula (1) or (2) shown below, alone or as a component in a mixture:



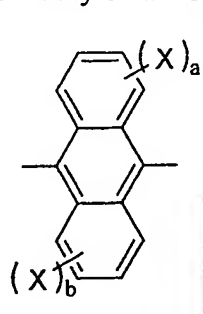
(In the formula, Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms; Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms; X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxy carbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group; a, b, and c are whole numbers of 0-4; and n is a whole number of 1-3; when n is 2 or more, the formula inside the parenthesis shown below may be the same or different:



Furthermore, the present invention provides anthracene derivatives represented by general formula (2) shown below:



(In the formula, Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms; Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms; X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxycarbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group; a, b, and c are whole numbers of 0-4; and n is a whole number of 1-3; when n is 2 or more, the formula inside the parenthesis shown below may be the same or different:



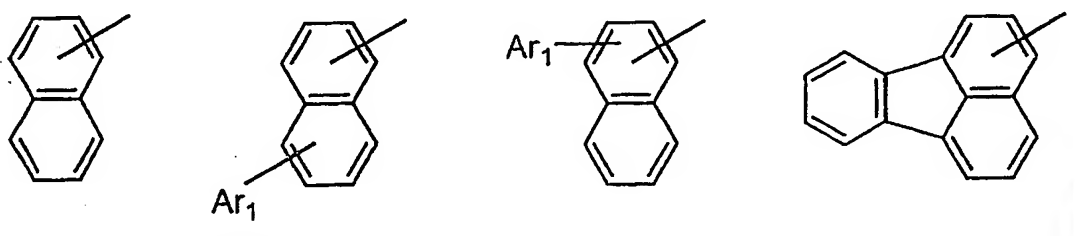
Best embodiments for practicing the invention

The present invention provides organic electroluminescence devices having at least one luminescence-layer-containing organic thin film layer sandwiched between the positive electrode and negative electrode, wherein at least one layer of the organic thin film layer contains the anthracene derivative represented by general formula (1) shown above, alone or as a component in a mixture.

In general formula (1), Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms.

The condensed aromatic group may be, e.g., a 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, etc.

The preferred condensed aromatic group of Ar can be selected from the general formulas given below:



(Ar<sub>1</sub> is an (un)substituted aromatic group of 6-50 nuclear carbon atoms.)

Ar<sub>1</sub> may be, e.g., a phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenyl group, 4''-t-butyl-p-terphenyl-4-yl group, etc.

In general formula (1), Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms. Such an aromatic group may be a phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, p-terphenyl-4-yl group, p-terphenyl-

3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenyl group, 4''-t-butyl-p-terphenyl-4-yl group, etc.

Of these, especially preferred are (un)substituted aromatic groups of 10 or more nuclear carbon atoms, such as a 1-naphthyl group, 2-naphthyl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, etc.

In general formula (1), X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxycarbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group.

Examples of the (un)substituted aromatic groups in X are the phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenyl group, 4''-t-butyl-p-terphenyl-4-yl group, etc.

Examples of the (un)substituted aromatic heterocyclic groups in X are the 1-pyrrolyl group, 2-pyrrolyl group, 3-pyrrolyl group, pyrazinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 2-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl

group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxaliny group, 5-quinoxaliny group, 6-quinoxaliny group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 9-carbazolyl group, 1-phenanthridiny group, 2-phenanthridiny group, 3-phenanthridiny group, 4-phenanthridiny group, 6-phenanthridiny group, 7-phenanthridiny group, 8-phenanthridiny group, 9-phenanthridiny group, 10-phenanthridiny group, 1-acridiny group, 2-acridiny group, 3-acridiny group, 4-acridiny group, 9-acridiny group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8-phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-phenaziny group, 2-phenaziny group, 1-phenothiaziny group, 2-phenothiaziny group, 3-phenothiaziny group, 4-phenothiaziny group, 10-phenothiaziny group, 1-phenoxaziny group, 2-phenoxaziny group, 3-phenoxaziny group, 4-phenoxaziny group, 10-phenoxaziny group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl, 3-furazany group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methylpyrrol-5-yl group, 3-methylpyrrol-1-yl group, 3-methylpyrrol-2-yl group, 3-methylpyrrol-4-yl group, 3-methylpyrrol-5-yl group, 2-t-butylpyrrol-4-yl group, 3-(2-phenylpropyl)pyrrole-1-yl group, 2-methyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-1-indolyl group, 2-t-butyl-3-indolyl group, 4-t-butyl-3-indolyl group, etc.

Examples of the (un)substituted alkyl group in X are the methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-



pentyl group, n-hexyl group n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxyisopropyl group, 2,3-dihydroxy-t-butyl group, 1,2,3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2,3-trichloropropyl group, bromomethyl group, 1-bromoethyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2,3-tribromopropyl group, iodomethyl group, 1-iodoethyl group, 2-iodoethyl group, 2-iodoisobutyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, 2,3-diiodo-t-butyl group, 1,2,3-triiodopropyl group, aminomethyl group, 1-aminoethyl group, 2-aminoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2,3-triaminopropyl group, cyanomethyl group, 1-cyanoethyl group, 2-cyanoethyl group, 2-cyanoisobutyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2,3-tricyanopropyl group, nitromethyl group, 1-nitroethyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group, 1,2,3-trinitropropyl group, cyclopropyl group, cyclobutyl group, cyclopentyl group, cyclohexyl group, 4-methylcyclohexyl group, 1-adamantyl group, 2-adamantyl group, 1-norbornyl group, 2-norbornyl group, etc.

The (un)substituted alkoxy group is a group represented by  $-OY$ , and examples of Y are the methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxyisopropyl group, 2,3-dihydroxy-t-butyl group, 1,2,3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2,3-trichloropropyl group, bromomethyl group, 1-bromoethyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2,3-tribromopropyl group, iodomethyl group, 1-iodoethyl group, 2-iodoethyl group, 2-iodoisobutyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, 2,3-diiodo-t-butyl group, 1,2,3-triiodopropyl group, aminomethyl group, 1-aminoethyl group, 2-aminoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2,3-triaminopropyl group, cyanomethyl group, 1-cyanoethyl group, 2-cyanoethyl group, 2-cyanoisobutyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2,3-tricyanopropyl group, nitromethyl group, 1-nitroethyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group, 1,2,3-trinitropropyl group, etc.

Examples of the (un)substituted aralkyl group are the benzyl group, 1-phenylethyl group, 2-phenylethyl group, 1-phenylisopropyl group, 2-phenylisopropyl group, phenyl-*t*-butyl group,  $\alpha$ -naphthylmethyl group, 1- $\alpha$ -naphthylethyl group, 2- $\alpha$ -naphthylethyl group, 1- $\alpha$ -naphthylisopropyl group, 2- $\alpha$ -naphthylisopropyl group,  $\beta$ -naphthylmethyl group, 1- $\beta$ -naphthylethyl group, 2- $\beta$ -naphthylethyl group, 1- $\beta$ -naphthylisopropyl group, 2- $\beta$ -naphthylisopropyl group, 1-pyrrolylmethyl group, 2-(1-pyrrolyl)ethyl group, *p*-methylbenzyl group, *m*-methylbenzyl group, *o*-methylbenzyl group, *p*-bromobenzyl group, *m*-bromobenzyl group, *o*-bromobenzyl group, *p*-iodobenzyl group, *m*-iodobenzyl group, *o*-iodobenzyl group, *p*-hydroxybenzyl group, *m*-hydroxybenzyl group, *o*-hydroxybenzyl group, *p*-aminobenzyl group, *m*-aminobenzyl group, *o*-aminobenzyl group, *p*-nitrobenzyl group, *m*-nitrobenzyl group, *o*-nitrobenzyl group, *p*-cyanobenzyl group, *m*-cyanobenzyl group, *o*-cyanobenzyl group, 1-hydroxy-2-phenylisopropyl group, 1-chloro-2-phenylisopropyl group, etc.

The (un)substituted aryloxy group is represented by  $-OY'$ , and examples of  $Y'$  are the phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, *p*-terphenyl-4-yl group, *p*-terphenyl-3-yl group, *p*-terphenyl-2-yl group, *m*-terphenyl-4-yl group, *m*-terphenyl-3-yl group, *m*-terphenyl-2-yl group, *o*-tolyl group, *m*-tolyl group, *p*-tolyl group, *p*-*t*-butylphenyl group, *p*-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenyl group, 4''-*t*-butyl-*p*-terphenyl-4-yl group, 1-pyrrolyl group, 2-pyrrolyl group, 3-pyrrolyl group, pyrazinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, quinolyl group, 2-quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxaliny group, 5-quinoxaliny group, 6-quinoxaliny group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3-phenanthridinyl group, 4-phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-

phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4-acridinyl group, 6-acridinyl group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8-phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-phenazinyl group, 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 3-phenothiazinyl group, 4-phenothiazinyl group, 10-phenothiazinyl group, 1-phenoxazinyl group, 2-phenoxazinyl group, 3-phenoxazinyl group, 4-phenoxazinyl group, 10-phenoxazinyl group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl, 3-furazanyl group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methylpyrrol-5-yl group, 3-methylpyrrol-1-yl group, 3-methylpyrrol-2-yl group, 3-methylpyrrol-4-yl group, 3-methylpyrrol-5-yl group, 2-t-butylpyrrol-4-yl group, 3-(2-phenylpropyl)pyrrole-1-yl group, 2-methyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-1-indolyl group, 2-t-butyl-3-indolyl group, 4-t-butyl-3-indolyl group, etc.

The (un)substituted arylthio group is represented by  $-SY''$ , and examples of  $Y''$  are the phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenyl group, 3-biphenyl group, 4-biphenyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-

terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl)phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenylyl group, 4''-t-butyl-p-terphenyl-4-yl group, 2-pyrrolyl group, 3-pyrrolyl group, pyrazinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, quinolyl group, 2-quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxalinyll group, 5-quinoxalinyll group, 6-quinoxalinyll group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3-phenanthridinyl group, 4-phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4-acridinyl group, 9-acridinyl group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8-phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-

phenazinyl group, 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 3-phenothiazinyl group, 4-phenothiazinyl group, 1-phenoxazinyl group, 2-phenoxazinyl group, 3-phenoxazinyl group, 4-phenoxazinyl group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl, 3-furazanyl group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methylpyrrol-5-yl group, 3-methylpyrrol-1-yl group, 3-methylpyrrol-2-yl group, 3-methylpyrrol-4-yl group, 3-methylpyrrol-5-yl group, 2-t-butylpyrrol-4-yl group, 3-(2-phenylpropyl)pyrrole-1-yl group, 2-methyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-1-indolyl group, 2-t-butyl-3-indolyl group, 4-t-butyl-3-indolyl group, etc.

The (un)substituted alkoxycarbonyl group is represented by  $-\text{COOZ}$ , and examples of Z are the methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxyisopropyl group, 2,3-dihydroxy-t-butyl group, 1,2,3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2,3-trichloropropyl group, bromomethyl group, 1-bromoethyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2,3-tribromopropyl group, iodomethyl group, 1-iodoethyl group, 2-iodoethyl group, 2-iodoisobutyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, 2,3-diiodo-t-butyl group, 1,2,3-triiodopropyl group, aminomethyl group, 1-aminoethyl group, 2-aminoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2,3-triaminopropyl group, cyanomethyl group, 1-cyanoethyl group, 2-cyanoethyl group, 2-cyanoisobutyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2,3-tricyanopropyl group, nitromethyl group, 1-nitroethyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group, 1,2,3-trinitropropyl group, etc.

Examples of the ring-forming divalent groups are the tetramethylene group, pentamethylene group, hexamethylene group, diphenylmethane-2,2'-di-yl group, diphenylethane-3,3'-di-yl group, diphenylpropane-4,4'-di-yl group, etc.

The halogen atom may be fluorine, chlorine, bromine, or iodine.

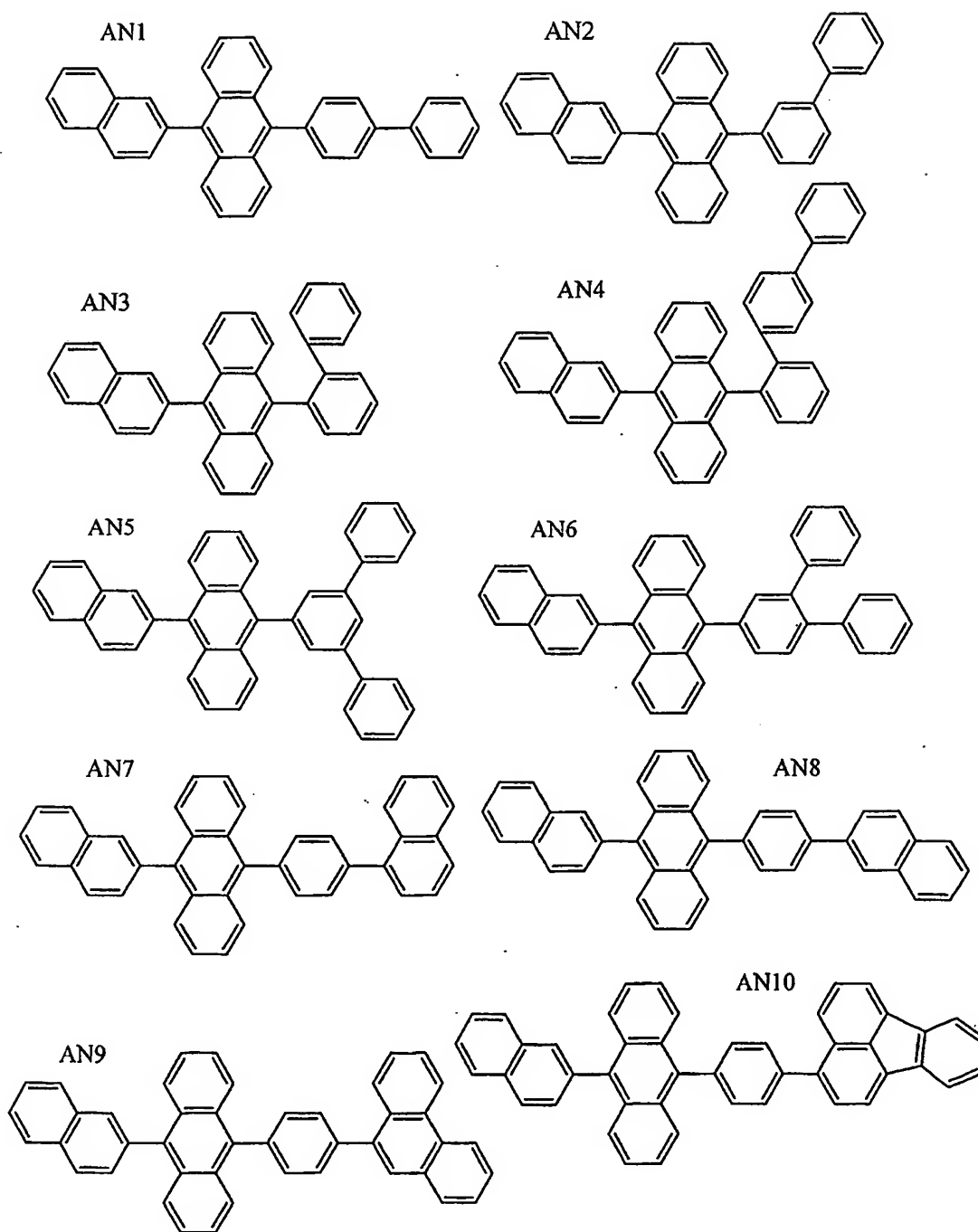
In general formula (1), a, b, and c independently represent whole numbers of 0-4, preferably 0-1.

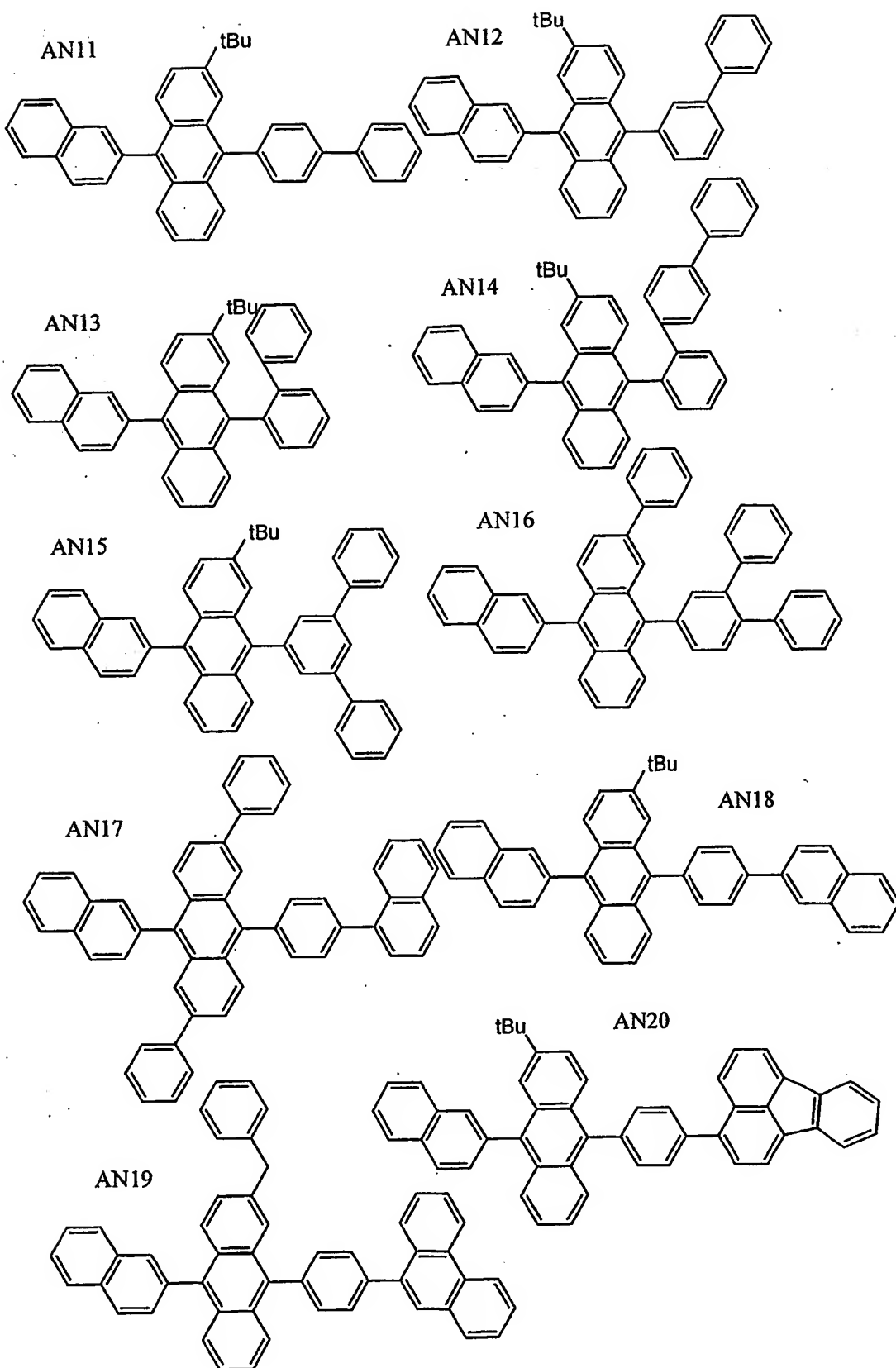
The n is a whole number of 1-3. When n is 2 or more, the groups in [ ] may be the same or different.

Substituents on the groups represented by the above Ar, Ar', and X may be a halogen atom, hydroxy group, nitro group, cyano group, alkyl group, aryl group, cycloalkyl group, alkoxy group, aromatic heterocyclic group, aralkyl group, aryloxy group, arylthio group, alkoxycarbonyl group, or carboxy group.

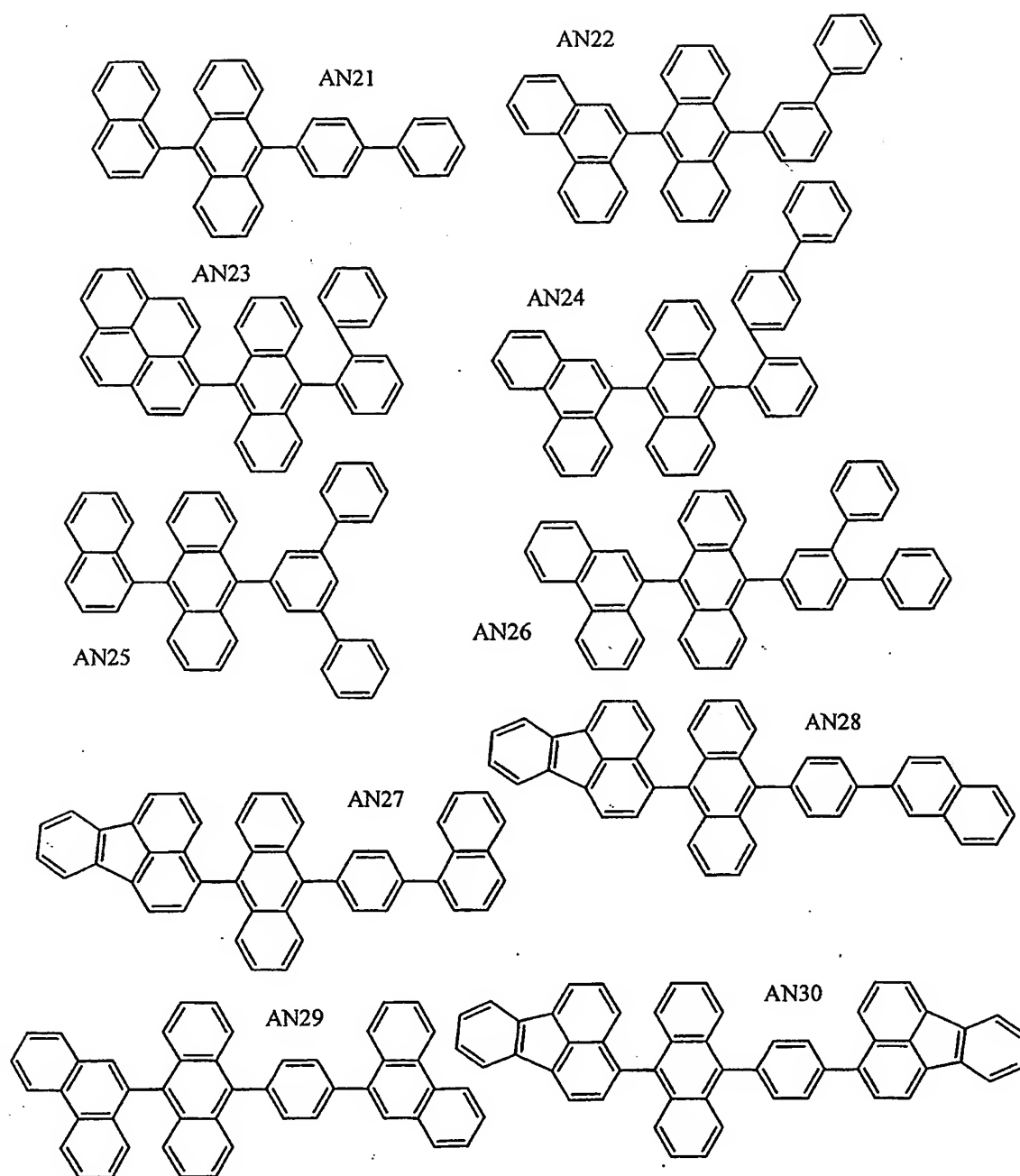
Specific examples of the anthracene derivative represented by general formula (1) of the present invention are, but not limited to, those shown below. Me is a methyl group and Bu is a butyl group.

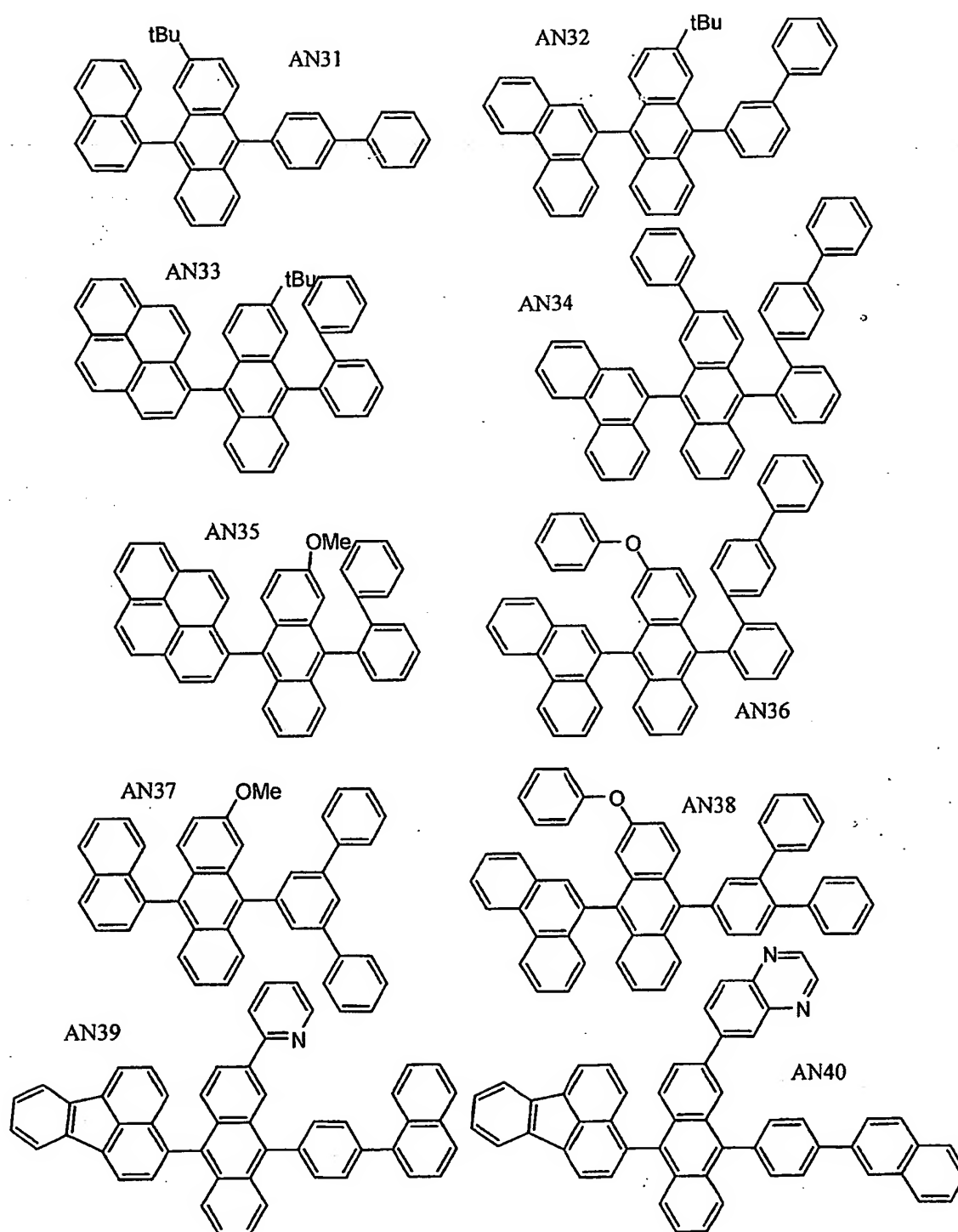
In the anthracene derivatives represented by general formula (1), both Ar and Ar' representing a naphthyl group and  $a = b = c = 0$  are most preferred.

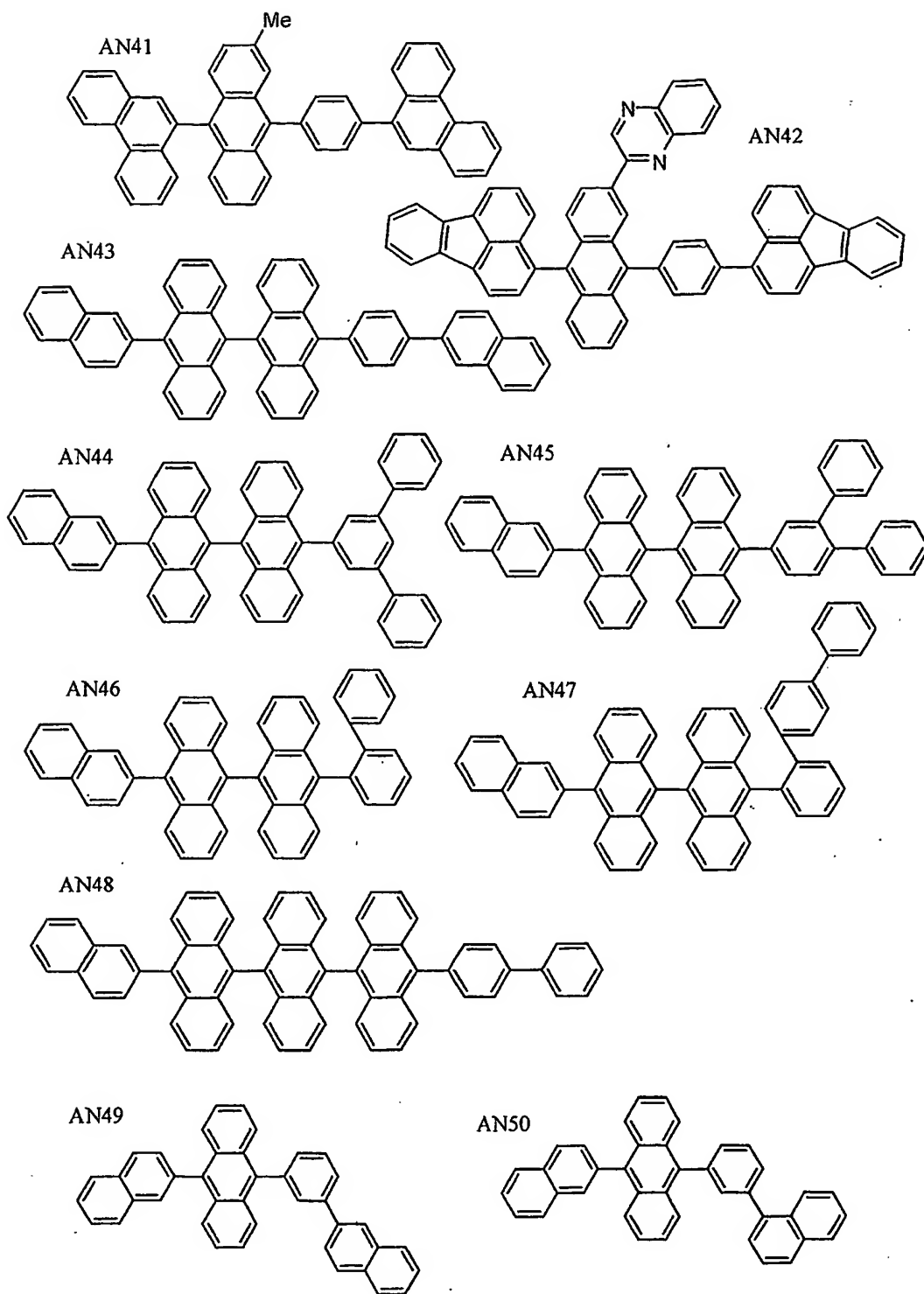




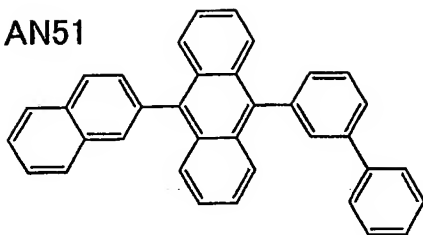




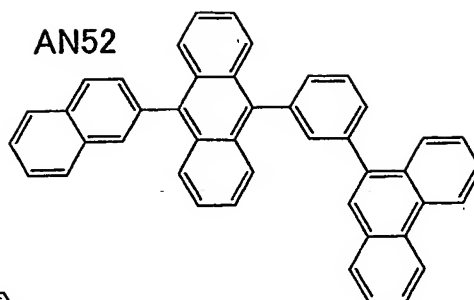




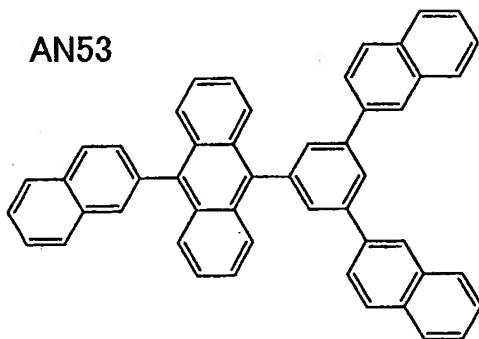
AN51



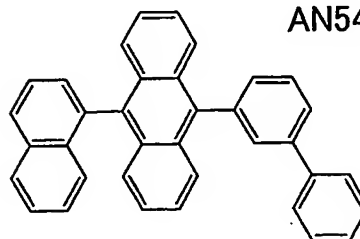
AN52



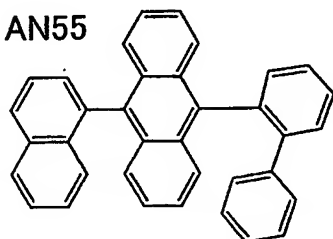
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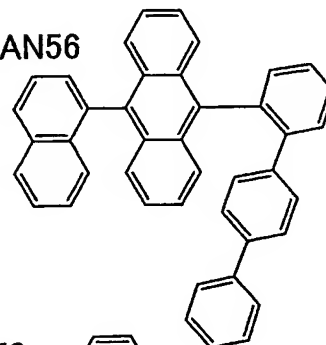
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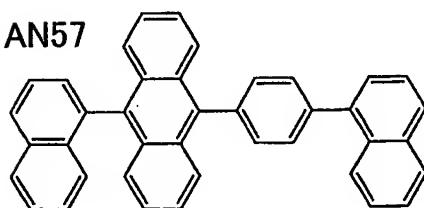
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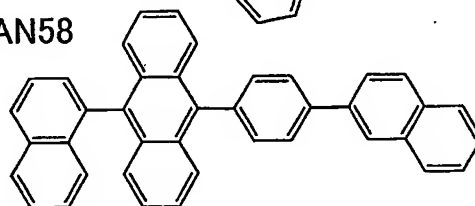
AN56



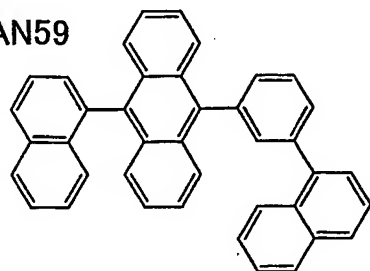
AN57



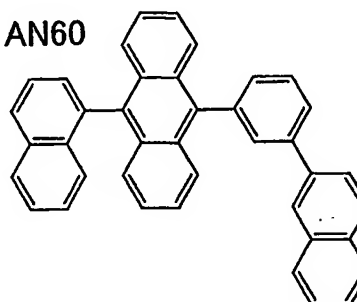
AN58



AN59



AN60



The anthracene derivatives represented by the above general formula (2) of the present invention, included in general formula (1), are novel compounds.

- In general formula (2), Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms.
- In general formula (2), Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms.
- In general formula (2), X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxycarbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group.
- The specific examples of groups represented by Ar, Ar', and X are similar to those given for general formula (1).

Substituents on the groups represented by the above Ar, Ar', and X may be a halogen atom, hydroxy group, nitro group, cyano group, alkyl group, aryl group, cycloalkyl group, alkoxy group, aromatic heterocyclic group, aralkyl group, aryloxy group, arylthio group, alkoxycarbonyl group, or carboxy group.

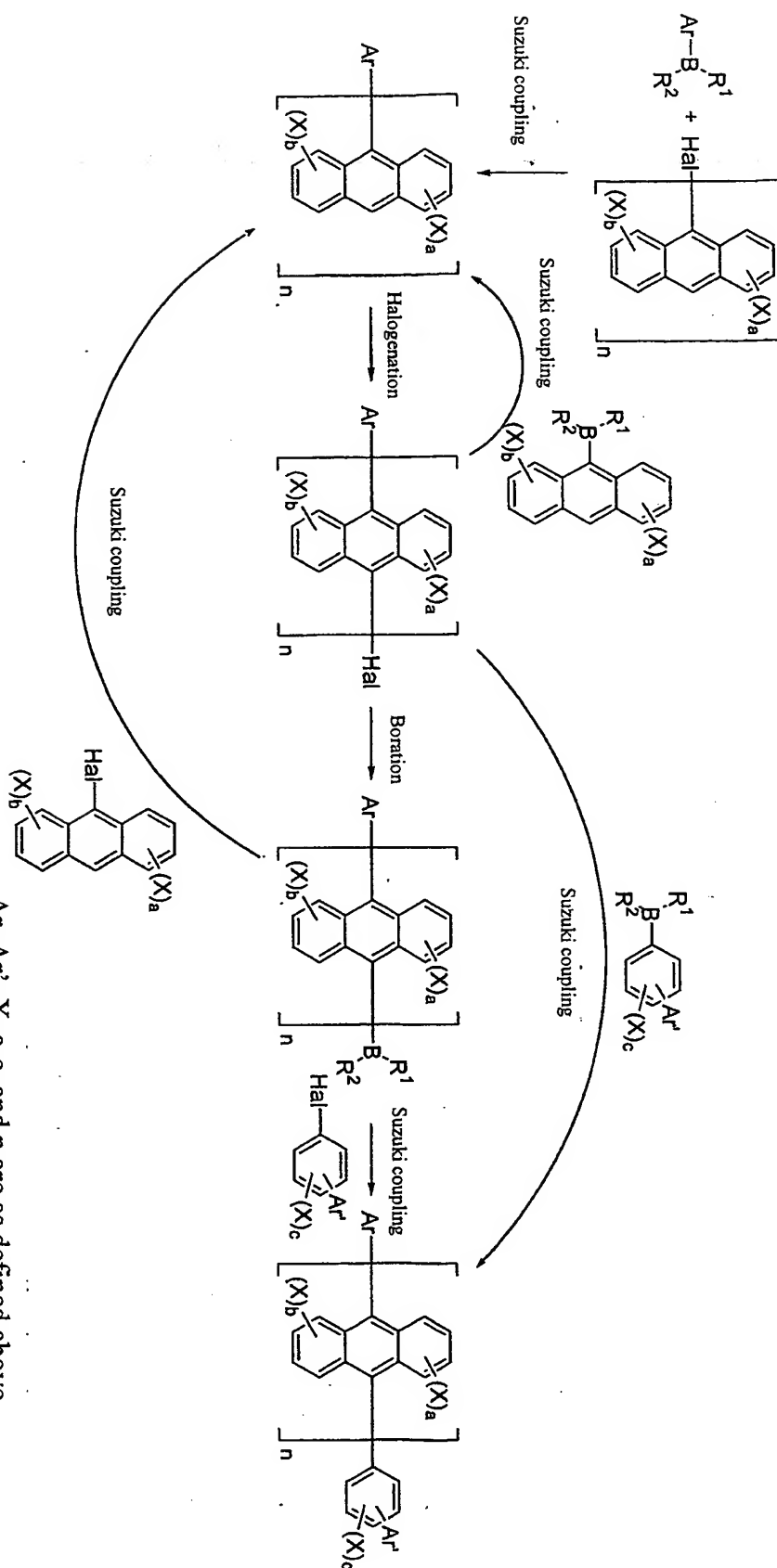
In general formula (2), a and b independently represent whole numbers of 0-4, preferably 0-1.

The n is a whole number of 1-3. When n is 2 or more, the groups in [ ] may be the same or different.

Specific examples of the anthracene derivatives represented by general formula (2) of the present invention may be, but not limited to, (AN1)-(AN4), AN7)-(AN14), (AN17)-(AN24), (AN27)-(AN36), (AN39)-(AN43), and (AN46)-(AN48) given as specific examples of general formula (1).

The anthracene derivatives represented by general formula (2) of the present invention are preferred for organic EL device materials.

The anthracene derivatives of general formula (1) or (2), used in the organic EL devices of the present invention, can be synthesized by an appropriate combination of Suzuki coupling reaction, halogenation, and boration reaction of a commercially available arylboronic acid or arylboronic acid synthesized by a known method and using halogenated anthracene derivatives as starting materials. The synthesis scheme is shown below.



Ar, Ar', X, a-c, and n are as defined above  
 $R^1$ ,  $R^2$ : alkoxy group with or without a hydroxy group  
 or substituent

There have been many reports of the Suzuki coupling reaction (Chem. Rev., Vol. 95, No. 7, 2457 (1995), etc.), and this reaction can be carried out under conditions described in such reports.

Usually, the reaction is carried out under ambient pressure in an inert atmosphere of nitrogen, argon, helium, etc., and under pressure if needed. The reaction temperature is in the range of 15-300°C, preferably 30-200°C.

The reaction solvents may be water, aromatic hydrocarbons such as benzene, toluene, xylene, etc.; ethers such as 1,2-dimethoxyethane, diethyl ether, methyl t-butyl ether, tetrahydrofuran, dioxane, etc.; saturated hydrocarbons such as pentane, hexane, heptane, octane, cyclohexane, etc.; halogen compounds such as dichloromethane, chloroform, carbon tetrachloride, 1,2-dichloroethane, 1,1,1-trichloroethane, etc.; nitriles such as acetonitrile, benzonitrile, etc.; esters such as ethyl acetate, methyl acetate, butyl acetate, etc.; amides such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylpyrrolidone, etc.; or mixtures thereof. Of these, preferred are toluene, 1,2-dimethoxyethane, dioxane, and water. The amount of solvent used is usually 3-50 times, preferably 4-20 times by weight of the arylboronic acid or its derivatives.

The base used in the reaction may be, e.g., sodium carbonate, potassium carbonate, sodium hydroxide, potassium hydroxide, sodium bicarbonate, potassium bicarbonate, magnesium carbonate, lithium carbonate, potassium fluoride, cesium fluoride, cesium chloride, cesium bromide, cesium carbonate, potassium phosphate, methoxysodium, t-butoxypotassium\*, t-butoxysodium\*, t-butoxylithium\*, etc., preferably sodium carbonate. The amount of such a base used is usually 0.7-10 molar equivalent, preferably 0.9-6 molar equivalent, with respect to the arylboronic acid or its derivatives.

The catalysts used in the reaction may be, e.g., palladium catalysts such as tetrakis(triphenylphosphine)palladium, dichlorobis(triphenylphosphine)palladium, dichloro[bis(diphenylphosphino)ethane]palladium, dichloro[bis(diphenylphosphino)propane]palladium, dichloro[bis(diphenylphosphino)butane]palladium, dichloro[bis(diphenylphosphino)ferrocene]palladium, etc.; nickel catalysts such as tetrakis(triphenylphosphine)nickel, dichlorobis(triphenylphosphine)nickel, dichloro[bis(diphenylphosphino)ethane]nickel, dichloro[bis(diphenylphosphino)propane]nickel, dichloro[bis(diphenylphosphino)butane]nickel, dichloro[bis(diphenylphosphino)ferrocene]nickel, etc., while tetrakis(triphenylphosphine)palladium is preferred. The amount of the catalyst used is

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\* [Butoxypotassium, etc., should possibly be "butylpotassium", etc.]

usually 0.001-1 molar equivalent, preferably 0.01-0.1 molar equivalent, with respect to the haloanthracene derivatives used.

The halogen in the haloanthracene derivatives may be, e.g., an iodine atom, bromine atom, chlorine atom, etc., preferably an iodine atom or bromine atom.

The halogenation agents used in the halogenation reaction may be, but not limited to, N-halosuccinimide. The amount of the halogenation agent used is usually 0.8-10 molar equivalent, preferably 1-5 molar equivalent, with respect to the anthracene derivatives.

The reaction is usually carried out in an inert solvent and in an inert atmosphere such as nitrogen, argon, helium, etc. The inert solvents that can be used are, e.g., N,N-dimethylformamide, N,N-dimethylacetamide, N-methylpyrrolidone, dimethyl sulfoxides, carbon tetrachloride, chlorobenzene, dichlorobenzene, nitrobenzene, toluene, xylene, methyl Cellosolve, ethyl Cellosolve, water, etc., preferably N,N-dimethylformamide and N-methylpyrrolidone. The amount of the solvents used is usually 3-50 times, preferably 5-20 times by weight of the anthracene derivatives. The reaction temperature is usually 0-200 °C, preferably 20-120 °C.

The boration reaction may be carried out in the usual manner (Nippon Kagakukai: Jikken Kagaku Goza, 4<sup>th</sup> ed., Vol. 24, pp. 61-90; J. Org. Chem., Vol. 60, 7508 (1995), etc.) For example, in the case of the reaction through the thiolation or Grignard reaction of haloanthracene derivatives, the reaction is usually carried out in an inert atmosphere such as nitrogen, argon, helium, etc., and in an inert solvent, e.g., saturated hydrocarbons such as pentane, hexane, heptane, octane, cyclohexane, etc.; ethers such as 1,2-dimethoxyethane, diethyl ether, methyl t-butyl ether, tetrahydrofuran, dioxane, etc.; aromatic hydrocarbons such as benzene, toluene, xylene, etc., or mixtures thereof, preferably diethyl ether and toluene. The amount of solvents used is usually 3-50 times, preferably 4-20 times the haloanthracene derivatives by weight.

The lithiation agents may be, e.g., alkylmetal reagents such as n-butyllithium, t-butyllithium, phenyllithium, methyllithium, etc.; amide bases such as lithium diisopropylamide, lithium bistrimethylsilylamide, etc., preferably n-butyllithium. The Grignard reagents can be prepared by reacting haloanthracene derivatives with metallic magnesium. The trialkyl borates that can be used as boration agents are, e.g., trimethyl borate, triethyl borate, triisopropyl borate, tributyl borate, etc., preferably trimethyl borate and triisopropyl borate.

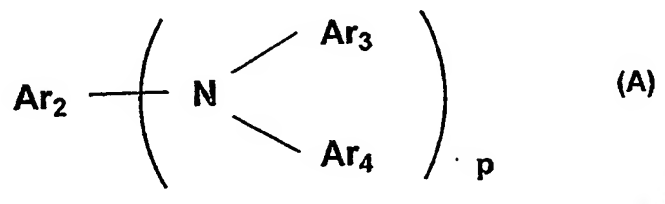
Usually, the amounts of the lithiation agents and metallic magnesium used are, respectively, 1-10 molar equivalent, preferably 1-2 molar equivalent with respect to the haloanthracene derivatives. The amount of trialkyl borate used is usually 1-10 molar equivalent, preferably 1-5 molar equivalent, with respect to the haloanthracene derivatives. The reaction temperature is usually -100 to 50 °C, preferably -75 to 10 °C.

In the organic EL devices of the present invention, preferably, the luminescence layer contains anthracene derivatives represented by general formula (1) or (2), as the main component.



Furthermore, in the organic EL devices of the present invention, preferably, the luminescence layer contains also an arylamine compound and/or styrylamine compound.

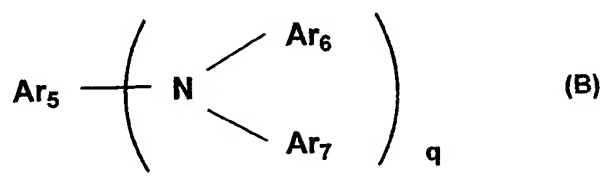
The styrylamine compounds are preferably those represented by general formula (A) shown below:



(In the formula, Ar<sub>2</sub> is a group chosen from a phenyl group, biphenyl group, terphenyl group, stilbene group, or distyrylaryl group; Ar<sub>3</sub> and Ar<sub>4</sub> independently represent a hydrogen atom or C<sub>6-20</sub> aromatic group; Ar<sub>2</sub>, Ar<sub>3</sub>, and Ar<sub>4</sub> may be substituted. The p is a whole number of 1-4. Further preferably, at least one of Ar<sub>3</sub> and Ar<sub>4</sub> is substituted by a styryl group.)

Here, the C<sub>6-20</sub> aromatic group may be phenyl group, naphthyl group, anthranil group, phenanthryl group, terphenyl group, etc.

Arylamine compounds are preferably those represented by general formula (B) shown below:



(In formula, Ar<sub>5</sub>-Ar<sub>7</sub> are (un)substituted aryl groups of 5-40 nuclear carbon atoms; q is a whole number of 1-4.)

The aryl groups of 5-40 nuclear carbon atoms are, e.g., the phenyl group, naphthyl group, anthranil group, phenanthryl group, pyrenyl group, koroniru [transliteration] group, biphenyl group, terphenyl group, pyrrolyl group, furanyl group, thiophenyl group, benzothiophenyl group, oxadiazolyl group, diphenylanthranil group, indolyl group, carbazolyl group, pyridyl group, benzoquinolyl, fluoranthenyl group, acenaphthofluoranthenyl group, stilbene group, etc. Preferred substituents for such aryl groups are the C<sub>1-6</sub> alkyl group (ethyl group, methyl group, i-propyl group, n-propyl group, s-butyl group, t-butyl group, pentyl group, hexyl group, cyclopentyl group, cyclohexyl group, etc.), C<sub>1-6</sub> alkoxy group (ethoxy group, methoxy group, i-propoxy group, n-propoxy group, s-butoxy group, t-butoxy group, pentoxy group, hexyloxy group, cyclopentoxy group, cyclohexyloxy group, etc.), aryl group of 5-40 nuclear atoms, amino group substituted by an aryl group of 5-40 nuclear atoms, ester group having an aryl group of 5-40 nuclear atoms, ester group having a C<sub>1-6</sub> alkyl group, cyano group, nitro group, halogen atom, etc.

Next, the constitution of the organic EL devices of the present invention is explained.

Main constitutions of the organic EL devices of the present invention are:

- (1) Positive electrode/luminescence layer/negative electrode
- (2) Positive electrode/positive hole injection layer/luminescence layer/negative electrode
- (3) Positive electrode/luminescence layer/electron injection layer/negative electrode
- (4) Positive electrode/positive hole injection layer/luminescence layer/electron injection layer/negative electrode
- (5) Positive electrode/organic semiconductor layer/luminescence layer/negative electrode
- (6) Positive electrode/organic semiconductor layer/electron barrier layer/luminescence layer/negative electrode
- (7) Positive electrode/organic semiconductor layer/luminescence layer/adhesion improving layer/negative electrode
- (8) Positive electrode/positive hole injection layer/positive hole transport layer/luminescence layer/electron injection layer/negative electrode
- (9) Positive electrode/insulation layer/luminescence layer/insulation layer/negative electrode
- (10) Positive electrode/inorganic semiconductor layer/insulation layer/luminescence layer/insulation layer/negative electrode
- (11) Positive electrode/organic semiconductor layer/insulation layer/luminescence layer/insulation layer/negative electrode
- (12) Positive electrode/insulation layer/positive hole injection layer/positive hole transport layer/luminescence layer/insulation layer/negative electrode
- (13) Positive electrode/insulation layer/positive hole injection layer/positive hole transport layer/luminescence layer/electron injection layer/negative electrode, etc.

Of these, the constitution (8) is preferably used, but one is not limited to it.

Such organic EL devices are usually prepared on a transparent substrate. Such a transparent substrate is a base supporting the organic EL device and should have a light transmittance above 50% for visible light in 400-700 nm range; a smooth substrate is further preferred.

Such transparent substrates are preferably glass plates and synthetic resin plates. Glass plates may be especially soda ash glass, glass containing barium and strontium, lead glass, aluminosilicate glass, borosilicate glass, barium borosilicate glass, quartz, etc. Synthetic resin plates may be plates made from polycarbonate resins, acrylic resins, polyethylene terephthalate resin, polyether sulfide resins, polysulfone resins, etc.

The positive electrode should be made from high-work-function (above 4 eV) metals, alloys, electrically conductive compounds, or mixtures thereof. Specific examples of such electrode materials are metals such as Au, etc., and electrically conductive material such as CuI, ITO (indium tin oxide),  $\text{SnO}_2$ , ZnO, In-Zn-O, etc. Such a positive electrode can be formed by

the thin film process by vapor deposition, sputtering, etc., of the electrode materials. When the light emitted from the luminescence layer is taken from the positive electrode, the positive electrode should have a light transmittance above 10%. The positive electrode sheet resistance should be below several hundreds of  $\Omega/\square$ . Furthermore, depending on the type of materials, usually, the positive electrode has a thickness of 10 nm~1  $\mu$ m, preferably 10-200 nm.

Next, the negative electrode should be made from low-work-function (below 4 eV) metals, alloys, electrically conductive compounds, and mixtures thereof. Specific examples of such electrode materials include sodium, sodium-potassium alloy, magnesium, lithium, magnesium-silver alloy, aluminum/aluminum oxide, Al/Li<sub>2</sub>O, Al/Li<sub>2</sub>O, Al/LiF, aluminum-lithium alloy, indium, rare earth metals, etc.

Such a negative electrode can be formed by the thin film process by vapor deposition, sputtering, etc., of the electrode materials.

When the light emitted from the luminescence layer is taken from the negative electrode, the negative electrode should have a light transmittance above 10%. The negative electrode sheet resistance should be below several hundreds of  $\Omega/\square$ . Furthermore, usually, the negative electrode has a thickness of 10 nm~1  $\mu$ m, preferably 10-200 nm.

In the organic EL devices of the present invention, preferably a chalcogenide layer, metal halide layer, or metal oxide layer (hereafter, called surface layer in some cases) is formed on the surface of at least one of the pair of electrodes prepared. More specifically, a chalcogenide (including oxide) layer of a metal such as silicon, aluminum, etc., is formed on the positive electrode surface at the luminescence layer side, and a metal halide layer or metal oxide layer being formed on the negative electrode surface at the luminescence layer side. By doing this, the operation can be stabilized.

The chalcogenides are preferably SiO<sub>x</sub> ( $1 \leq x < 2$ ), AlO<sub>x</sub> ( $1 \leq x \leq 1.5$ ), SiON, SiAlON, etc. Metal halides may be LiF, MgF<sub>2</sub>, CaF<sub>2</sub>, rare earth metal fluorides, etc. Metal oxides may be Cs<sub>2</sub>O, Li<sub>2</sub>O, MgO, SrO, BaO, CaO, etc.

In the organic EL devices of the present invention, it is also preferred that a mixed area of an electron conducting compound and reductive dopant or a mixed area of a positive hole conducting compound and oxidative dopant is installed on the surface of at least one of the pair of electrodes prepared above. By doing this, the electron conducting compound is reduced to an anion, making the mixed area easier to inject and to conduct electrons to the luminescence layer. The positive hole conducting compound is oxidized to a cation, making the mixed area easier to inject and to conduct the positive holes to the luminescence layer. Preferred oxidative dopants are various Lewis acids and acceptor compounds. Preferred reductive dopants are alkali metals, alkali metal compounds, alkaline earth metals, rare earth metals, and compounds thereof.

In the organic EL devices of the present invention, the luminescence layer has:

- (1) an injection function: capable of injecting positive holes from the positive electrode or positive hole injection layer in the presence of an electric field;
- (2) a transport function: capable of moving the injected charge (electron and positive hole) by the force of the electric field;
- (3) a luminescence function: providing a field of recombination of the electron and positive hole, leading to light emission.

Such a luminescence layer can be formed by known methods such as vapor deposition, spin coating, LB process, etc. Molecular deposition films are especially preferred for the luminescence layer. Such molecular deposition films may be thin films deposited from gas-phase compounds or films formed by solidification of compounds in the solution or liquid state. Usually, such molecular deposition films are different from thin films formed by the LB process (molecular accumulation films) in an aggregate structure or high-order structure, and with respect to functions caused by such differences.

As disclosed in Japanese Kokai Patent No. SHO 57[1982]-51781, the luminescence layer can also be formed by forming thin films by the spin coating, etc., of a solution containing binders such as resins, etc., and the [luminescent] material compounds.

In the present invention, within the scope of the present invention, if desired, the luminescence layer may also contain known luminescence materials other than those of the present invention. Furthermore, the luminescence layer containing the luminescent material of the present invention may be laminated with a luminescent layer containing other known luminescent material.

The positive hole injection-transport layer aids positive hole injection into the luminescence layer and transports it to the luminescence region; the positive hole mobility is high and the ionization energy is small, below 5.5 eV. For such a positive hole injection-transport layer, materials capable of transporting the positive hole to the luminescent layer at a low electric field strength are needed. For example, preferred material should have at least  $10^{-6}$  cm<sup>2</sup>/V-sec of positive hole mobility at an electric field of  $10^4 \sim 10^6$  V/cm. Such materials can be selected from those commonly used as positive charge transport materials in light conducting materials and those used in positive hole injection layers of organic EL devices.

Such a positive hole injection-transport layer may be formed by the thin film process by the usual methods such as vacuum vapor deposition, spin coating, casting, LB process, etc. of the positive hole injection-transport materials. The film thickness of the positive hole injection-transport layer is usually, but not limited to, 5 nm-5  $\mu$ m.

The electron injection-transport layer aids the injection of electrons into the luminescence layer and transports it to the luminescence region, with electron mobility being high. The adhesion improving layer is made of a material having an especially good adhesion to the

negative electrode in the electron injection layer. Materials used in the electron injection layer are preferably metal complexes of 8-hydroxyquinoline or its derivatives. Specific examples of the metal complexes of 8-hydroxyquinoline or its derivatives include metal chelate oxinoid compounds containing a chelate of oxine (in general, 8-quinolinol or 8-hydroxyquinoline). For example, tris(8-quinolinol)aluminum can be used for the electron injection material.

Furthermore, in general, with the application of an electric field to ultra-thin films, pixel defects are easily formed by leaking and short [circuit]. To prevent this, a thin insulation film layer can be inserted between the electrodes.

The materials used for the insulation layer are, e.g., aluminum oxide, lithium fluoride, lithium oxide, cesium fluoride, cesium oxide, magnesium oxide, magnesium fluoride, calcium oxide, calcium fluoride, aluminum nitride, titanium oxide, silicon oxide, germanium oxide, silicon nitride, boron nitride, molybdenum oxide, ruthenium oxide, vanadium oxide, etc., mixtures, and laminates thereof.

The organic EL devices of the present invention may be prepared using such materials by methods described above by forming a positive electrode, luminescence layer, positive hole injection layer if needed, electron injection layer if needed, and finally a negative electrode. Organic EL devices can also be prepared in reverse order from the negative electrode to the positive electrode.

Next, an example of making an organic EL device with the installation of a positive electrode/positive hole injection layer/luminescence layer/electron injection layer/negative electrode, in that order, on a transparent substrate is explained.

First, vapor deposition or the sputtering process is used for forming a thin film from positive electrode material to a thickness below 1  $\mu\text{m}$ , preferably 10-200 nm, on an appropriate transparent substrate. Next, a positive hole injection layer is installed on the positive electrode. The positive injection layer can be formed by vacuum vapor deposition, spin coating, casting, LB process, etc., as described above, while the vacuum vapor deposition process is preferred for the ease of obtaining uniform films and no pinholes. In forming a positive hole injection layer by the vacuum vapor deposition process, the vapor deposition conditions may vary according to the compounds used (positive hole injection layer material), crystal structure, and recombination structure of the desired positive hole injection layer, etc. In general, the vapor deposition source temperature is 50-450°C, vacuum is  $10^{-7}$ ~ $10^{-3}$  torr, vapor deposition rate is 0.01-50 nm/sec, substrate temperature is -50 to 300°C, and film thickness is 5 nm-5  $\mu\text{m}$ .

Next, the luminescence layer is installed on the positive hole injection layer. Also, this luminescence layer can be formed using the luminescent materials of the present invention by vacuum vapor deposition, sputtering, spin coating, casting, etc., for forming the luminescent material into a thin film, while the vacuum vapor deposition process is preferred for the ease of

uniform film formation and no pinhole formation. The vapor deposition conditions for forming a luminescent layer by vacuum vapor deposition may vary according to the compounds used, while they can be selected from conditions similar to those of the positive hole injection layer formation. The film thickness should be in the 10-40 nm range.

Next, an electron injection layer is installed on the luminescent layer. Also, in this case, similarly as in the positive hole injection layer and luminescent layer, the vacuum vapor deposition process is preferred for obtaining uniform films. Vapor deposition conditions can be selected from those used for the positive hole injection layer and luminescent layer.

Finally, lamination of the negative electrode produces an organic EL device. The metal negative electrode can be formed by vapor deposition or sputtering. However, the vacuum vapor deposition process is preferred for protection of the underlying organic layer from damage during film formation.

It is preferred that such organic EL devices are prepared at once from the positive electrode to negative electrode with one vacuum operation.

With dc-voltage application to this organic EL device, positive electrode +, and negative electrode -, light emission is observed at a voltage of 3-40 V. When voltage is applied at reverse polarity, current does not flow, with no light emission at all. Furthermore, when an ac voltage is applied, only with polarity of positive electrode + and negative electrode -, uniform light emission is observed. In this case, the ac applied may be in any wave form.

Next, the present invention is explained in further detail with examples. However, the present invention is not limited to such examples.

#### Synthesis Example 1

##### Synthesis of 10-(2-naphthyl)anthracene-9-boronic acid

In a 20-L flask under an Ar atmosphere, a solution of 549 g of 2-naphthaleneboronic acid (product of Tokyo Kasei Co.), 684 g of 9-bromoanthracene (product of Tokyo Kasei Co.), 61.5 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 4.9 L of toluene (product of Hiroshima Wako Co.), and 845.9 g of sodium carbonate (product of Hiroshima Wako Co.) in 4.9 L of water was heated under stirring under reflux for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and recrystallized from toluene to obtain 751 g of crystals.

In a 20-L flask under an Ar atmosphere, a mixture of 588 g of the above crystals, 4.5 L of dehydrated ether (product of Hiroshima Wako Co.), and 4.5 L of dehydrated toluene (product of Hiroshima Wako Co.) was cooled to -64°C using dry ice, treated over a period of 30 min with 1.2 L of 1.6M butyllithium/hexane solution (product of Hiroshima Wako Co.), allowed to react at -64°C for 2 h, treated over a period of 20 min with 866 g of triisopropyl boronate (product of

Tokyo Kasei Co.), allowed to return to room temperature, stirred for 12 h, cooled with ice, treated with 4 L of 2N hydrochloric acid at a temperature below 10°C, and treated with 1 L of toluene, followed by liquid separation, drying over sodium sulfate, concentration in vacuo, addition of hexane, and filtering out the crystals precipitated. The product was dissolved in 5 L of THF (tetrahydrofuran), treated with 500 mL of concentrated hydrochloric acid and 5 g of tetrabutylammonium bromide, stirred for 12 h, and filtered. The filtered crystals were dried to obtain 431 g of crystals.

The FD-MS (field desorption mass analysis) of this compound gave  $m/z = 348$  to  $C_{24}H_{17}BO_2 = 348$ , confirming this compound as 10-(2-naphthyl)anthracene-9-boronic acid (yield: 47%).

### Synthesis Example 2

#### Synthesis of 2-(4-bromophenyl)naphthalene

In a 300 mL-flask under an Ar atmosphere, a solution of 7.1 g of 2-naphthaleneboronic acid (product of Tokyo Kasei Co.), 12.9 g of 4-iodobromobenzene (product of Tokyo Kasei Co.), 0.6 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), and 12.7 g of sodium carbonate (product of Hiroshima Wako Co.) in 60 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and filtered. The crystals filtered were recrystallized from toluene to obtain 9.0 g of crystals.

The FD-MS of this compound gave  $m/z = 284, 282$  to  $C_{16}H_{11}Br = 283$ , confirming the product as 2-(4-bromophenyl)naphthalene (yield: 77%).

### Synthesis Example 3

#### Synthesis of 3-(4-bromophenyl)fluoranthene

In a 500-mL flask under an Ar atmosphere, a mixture of 62 g of fluoranthene and 250 mL of DMF (product of Hiroshima Wako Co.) was heated to 80°C for dissolution, treated with 60 g of N-bromosuccinimide (product of Hiroshima Wako Co.) at 50°C, stirred for 2 h, treated with 500 mL of purified water, and the crystals precipitated were filtered out and purified by column chromatography to obtain 10.5 g of crystals.

In a 500-mL flask under an Ar atmosphere, a mixture of 10.0 g of the above crystals, 120 mL of dehydrated ether (product of Hiroshima Wako Co.) and 120 mL of dehydrated toluene (product of Hiroshima Wako Co.) was cooled to -64°C using dry ice, treated over a period of 30 min with 25 mL of 1.6 M butyllithium/hexane solution (product of Hiroshima Wako Co.), allowed to react at -64°C for 2 h, treated with 8 g of tiisopropyl brononate (product of Tokyo Kasei Co.) over a period of 20 min, allowed to return to room temperature, stirred for 12 h, cooled with ice, treated with 100 mL of 2N hydrochloric acid at a temperature below 10°C, and

treated with 25 mL of toluene, followed by phase separation, drying over sodium sulfate, concentration in vacuo, addition of hexane, and filtering the crystals precipitated. The filtered crystals were dissolved in 120 mL of THF, treated with 15 mL of concentrated hydrochloric acid and 0.15 g of tetrabutylammonium bromide, stirred for 12 h, and the crystals precipitated were filtered out and dried to obtain 7.0 g of 3-fluorantheneboronic acid crystals.

In a 300-mL flask under an Ar atmosphere, a solution of 7.0 g of the above crystals (product of Tokyo Kasei Co.), 9.0 g of 4-iodobromobenzene (product of Tokyo Kasei Co.), 0.6 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), and 12.7 g of sodium carbonate (product of Hiroshima Wako Co.) in 60 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and recrystallized from toluene to obtain 6.4 g of crystals.

The FD-MS of this compound gave  $m/z = 358$  to  $C_{22}H_{15}Br = 357$ , confirming this compound as 3-(4-bromophenyl)fluoranthene (yield: 6%).

#### Synthesis Example 4

##### Synthesis of 10-(3-fluoranthenyl)anthracene-9-boronic acid

In a 300-mL flask under an Ar atmosphere, a solution of 7.85 g of 3-fluorantheneboronic acid, 6.84 g of 9-bromoanthracene (product of Tokyo Kasei Co.), 0.6 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 50 mL of toluene, and 8.5 g of sodium carbonate (product of Hiroshima Wako Co.) in 50 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and recrystallized from toluene to obtain 4.6 g of crystals.

In a 300-mL flask under an Ar atmosphere, a mixture of 4.5 g of the above crystals and 100 mL of dehydrated DMF (product of Hiroshima Wako Co.) was heated to 80°C for dissolution, treated with 2.3 g of N-bromosuccinimide (product of Hiroshima Wako Co.) at 50°C, stirred for 2 h, treated with 200 mL of purified water, and the crystals precipitated were filtered out and recrystallized from toluene to obtain 4.5 g of crystals.

In a 300-mL flask under Ar atmosphere, a mixture of 4.5 g of the above crystals, 50 mL of dehydrated ether (product of Hiroshima Wako Co.) and 50 mL of dehydrated toluene (product of Hiroshima Wako Co.) was cooled to -64°C with dry ice, treated dropwise with 7 mL of 1.6M butyllithium/hexane solution (product of Hiroshima Wako Co.) over a period of 30 min, allowed to react at -64°C for 2 h, treated dropwise with 5.6 g of triisopropyl boronate (product of Tokyo Kasei Co.) over a period of 20 min, allowed to return to room temperature, stirred for 12 h, cooled with ice, treated with 40 mL of 2N hydrochloric acid at a temperature below 10°C, and treated with 10 mL of toluene. After phase separation, the toluene solution was dried over sodium sulfate, concentrated in vacuo, treated with hexane, and the crystals precipitated were



filtered out, dissolved in 50 mL of THF, treated with 5 mL of concentrated hydrochloric acid and 0.1 g of tetrabutylammonium bromide, stirred for 12 h, and the crystals precipitated were filtered out and dried to obtain 3.6 g of crystals.

The FD-MS of this compound gave  $m/z = 422$  to  $C_{30}H_{19}BO_2 = 422$ , confirming this compound as 10-(3-fluoranthenyl)anthracene-9-boronic acid (yield: 32%).

#### Synthesis Example 5

##### Synthesis of 1-(4-bromophenyl)naphthalene

Synthesis Example 2 was repeated using 1-naphthaleneboronic acid in place of 2-naphthaleneboronic acid to obtain 29.9 g of colorless oil.

The FD-MS of this compound gave  $m/z = 284, 282$  to  $C_{16}H_{11}Br = 283$ , confirming this compound as 1-(4-bromophenyl)naphthalene (yield: 88%).

#### Synthesis Example 6

##### Synthesis of 2-(3-bromophenyl)naphthalene

Synthesis Example 2 was repeated using 3-iodobromobenzene in place of 4-iodobromobenzene to obtain 20.1 g of colorless oil.

The FD-MS of this compound gave  $m/z = 284, 282$  to  $C_{16}H_{11}Br = 283$ , confirming this compound as 1-(3-bromophenyl)naphthalene (yield: 75%).

#### Application Example 1

##### Synthesis of compound (AN8)

In a 300-mL flask under an Ar atmosphere, a solution of 5.98 g of the 10-(2-naphthyl)anthracene-9-boronic acid obtained in Synthesis Example 1, 4.05 g of 2-(4-bromophenyl)naphthalene obtained in Synthesis Example 2, 0.33 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 60 mL of 1,2-dimethoxyethane (product of Hiroshima Wako Co.), and 4.55 g of sodium carbonate (product of Hiroshima Wako Co.) in 21 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and purified by column chromatography to obtain 3.4 g of light yellow solid.

The FD-MS of this compound gave  $m/z = 506$  to  $C_{40}H_{26} = 506$ , confirming this compound as AN8 (yield: 47%).

#### Application Example 2

##### Synthesis of compound (AN10)

In a 300-mL flask under an Ar atmosphere, a solution of 5.98 g of the 10-(2-naphthyl)anthracene-9-boronic acid obtained in Synthesis Example 1, 5.13 g of 3-(4-bromophenyl)fluoranthene obtained in Synthesis Example 3, 0.33 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 60 mL of 1,2-dimethoxyethane, and 4.55 g of sodium carbonate (product of Hiroshima Wako Co.) in 21 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and purified by column chromatography to obtain 3.3 g of light yellow solid.

The FD-MS of this compound gave  $m/z = 580$  to  $C_{46}H_{28} = 580$ , confirming this compound as AN10 (yield: 40%).

#### Application Example 3

##### Synthesis of compound (AN28)

In a 300-mL flask under an Ar atmosphere, a solution of 7.24 g of the 10-(3-fluoranthenyl)anthracene-9-boronic acid obtained in Synthesis Example 4, 4.05 g of the 2-(4-bromophenyl)naphthalene obtained in Synthesis Example 2, 0.33 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 60 mL of 1,2-dimethoxyethane (product of Hiroshima Wako Co.), and 4.55 g of sodium carbonate (Hiroshima Wako Co.) in 21 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and purified by column chromatography to obtain 3.6 g of light yellow solid.

The FD-MS of this compound gave  $m/z = 580$  to  $C_{46}H_{28} = 580$ , confirming this compound as AN28 (yield: 43%).

#### Application Example 4

##### Synthesis of compound (AN30)

In a 300-mL flask under an Ar atmosphere, a solution of 7.24 g of the 10-(3-fluoranthenyl)anthracene-9-boronic acid obtained in Synthesis Example 4, 5.13 g of the 3-(4-bromophenyl)fluoranthene obtained in Synthesis Example 3, 0.33 g of tetrakis(triphenylphosphine)palladium(0) (product of Tokyo Kasei Co.), 60 mL of 1,2-dimethoxyethane (product of Hiroshima Wako Co.), and 4.55 g of sodium carbonate (Hiroshima Wako Co.) in 21 mL of water was heated under reflux under stirring for 24 h, allowed to cool to room temperature, and the crystals precipitated were filtered out and purified by column chromatography to obtain 3.1 g of a light yellow solid.

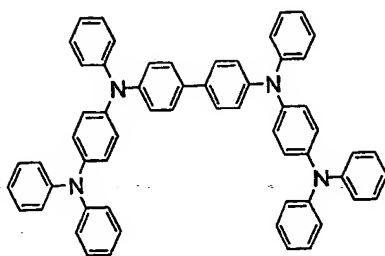
The FD-MS of this compound gave  $m/z = 654$  to  $C_{52}H_{30} = 654$ , confirming this compound as AN30 (yield: 33%).

### Application Example 5

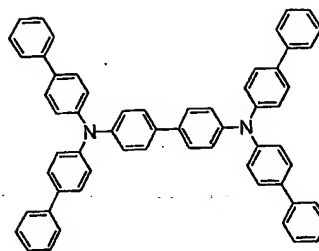
#### Manufacture of organic EL device

An ITO transparent electrode-fitted glass substrate (product of Diomatic Co.) of 25 mm x 75 mm x 1.1 mm (thickness) was cleaned by ultrasonic waves in isopropyl alcohol for 5 min, then by UV-ozone for 30 min. The cleaned transparent electrode line-fitted glass substrate was loaded in the substrate holder of a vacuum vapor deposition device. A 60-nm-thick N,N'-bis(N,N'-diphenyl-4-aminophenyl)-N,N-diphenyl-4,4'-diamino-1,1'-biphenyl film (hereafter referred to as TPD232 film) was formed on the transparent electrode side, covering the transparent electrode. This TPD232 film is the positive hole injection layer. A 20-nm-thick N,N,N',N'-tetra(4-biphenyl)-diaminobiphenylene film (hereafter referred to TBDB) was then formed on the TPD232 film. This film is the positive hole transport layer. A 40-nm-thick (AN8) film was formed as luminescent material on the TBDB film by vapor deposition. At the same time, the styryl-group-containing amine compound (D1) was vapor-deposited as luminescent material at AN8:D1 = 40:2 by weight. This film functions as the luminescent layer. A 10-nm-thick Alq film was then formed on the above film. This functions as an electron injection layer. Binary vapor deposition of the reductive dopant Li (Li source: product of Saes Target Co.) and Alq produced an Alq:Li film (film thickness: 10 nm) as an electron injection layer (negative electrode). Metallic Al was vapor deposited on the Alq:Li film to form a metallic negative electrode to obtain an organic EL device.

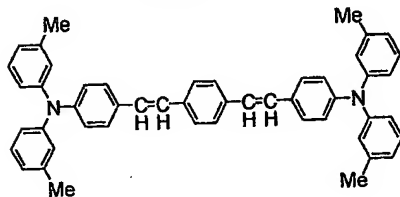
The organic EL device obtained was measured for luminescence efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.



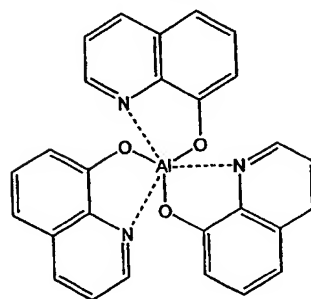
TPD232



TBDB



D 1



Alq

### Application Examples 6-8

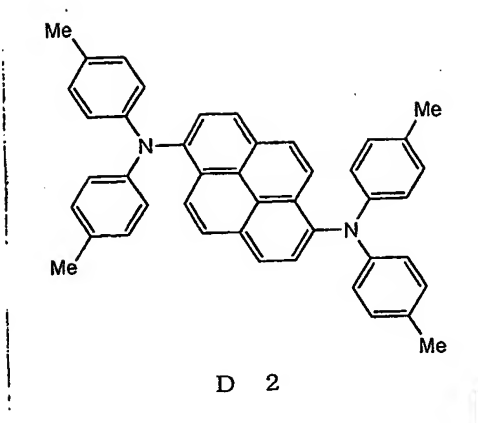
#### Manufacture of organic EL devices

Application Example 5 was repeated using the luminescent materials given in Table 1 in place of AN8 to obtain organic EL devices, which were measured for luminescence efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.

### Application Example 9

#### Manufacture of organic EL device

Application Example 5 was repeated using the aromatic amine (D2) in place of the styryl-group-containing amine compound (D1) to obtain an organic EL device, which was then measured for luminescence efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.



#### Application Example 10

##### Synthesis of compound (AN5)

Application Example 1 was repeated using 3,5-diphenylbromobenzene in place of 2-(4-bromophenyl)naphthalene to obtain 5.6 g of light yellow solid.

The FD-MS of this compound gave  $m/z = 532$  to  $C_{42}H_{28}Br = 532$ , confirming this compound as AN5 (yield: 45%).

#### Application Example 11

##### Synthesis of compound (AN7)

Application Example 1 was repeated using 1-(4-bromophenyl)naphthalene in place of 2-(4-bromophenyl)naphthalene to obtain 7.8 g of a light yellow solid.

The FD-MS of this compound gave  $m/z = 506$  to  $C_{40}H_{26}Br = 506$ , confirming this compound as AN7 (yield: 54%).

#### Application Example 12

##### Synthesis of compound (AN49)

Application Example 1 was repeated using 2-(3-bromophenyl)naphthalene in place of 2-(4-bromophenyl)naphthalene to obtain 6.9 g of light yellow solid.

The FD-MS of this compound gave  $m/z = 506$  to  $C_{40}H_{26}Br = 506$ , confirming this compound as AN7 (yield: 52%).

#### Application Examples 13-15

##### Manufacture of organic EL devices

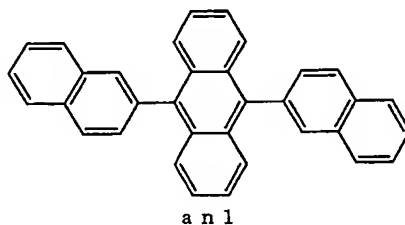
Application Example 5 was repeated using luminescent compounds described in Table 1 in place of AN8 to obtain organic EL devices, which were then measured for luminescence

efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.

#### Comparative Example 1

##### Manufacture of organic EL device

Application Example 5 was repeated using an1 as the luminescent material in place of AN8 to obtain an organic EL device, which was then measured for luminescence efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.



#### Comparative Example 2

##### Manufacture of organic EL device

Application Example 5 was repeated using an2 as the luminescent material in place of AN8 to obtain an organic EL device, which was then measured for luminescence efficiency and half-life under normal use at an initial brightness of 1000nit. Results are given in Table 1.

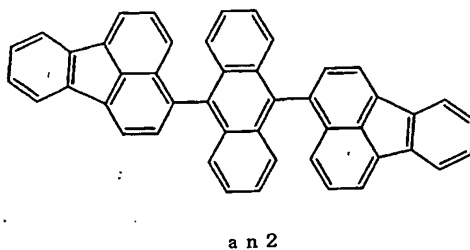


Table 1

	Fluorescent layer compound	Luminescence efficiency	Half-life	Luminescence color
		(cd/A)	(hours)	
AA5	AN8/D1	11.2	4200	Blue
AA6	AN10/D1	11.0	4000	Blue
AA7	AN28/D1	10.9	3700	Blue
AA8	AN30/D1	10.8	3700	Blue
AA9	AN8/D2	10.6	3200	Blue
AA13	AN5/D1	11.0	2200	Blue
AA14	AN7/D1	11.3	4500	Blue
AA15	AN49/D1	11.3	4500	Blue
CC1	an/D1	9.0	2200	Blue
CC2	an2/D1	8.8	1100	Blue

AA: Application Example

CC: Comparative Example

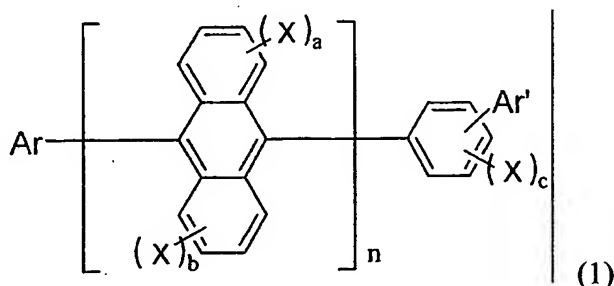
As shown in Table 1, the organic EL devices of Application Examples 5-9 and 13-15 have a high luminescence efficiency and very long service life, while the organic EL devices of Comparative Example 1 and 2 have a low luminescence efficiency and short service life.

#### Industrial application possibilities

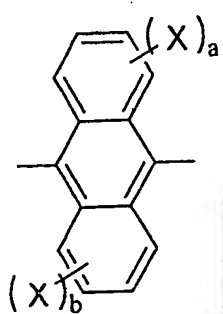
As explained in detail above, the organic EL devices of the present invention and the organic EL devices using the anthracene compounds of the present invention have a high luminescence efficiency and long service life, thus they are useful for organic EL devices expected for long-term continuous use.

### Claims

1. Organic electroluminescence device having at least one luminescence-layer-containing organic thin film layer sandwiched between the anode and cathode, wherein at least one layer of the organic thin film layer contains the anthracene derivative represented by general formula (1) shown below, alone or as a component in a mixture:

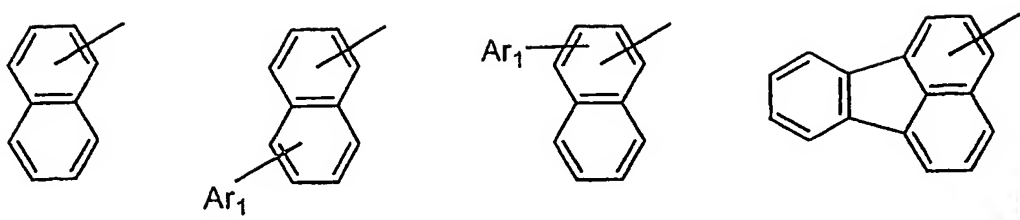


(in the formula, Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms; Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms; X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxycarbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group; a, b, and c are whole numbers of 0-4; and n is a whole number of 1-3; when n is 2 or more, the formula inside the parenthesis shown below may be the same or different:



2. The organic electroluminescence device described in Claim 1, wherein Ar in general formula (1) is selected from the general formulas given below:





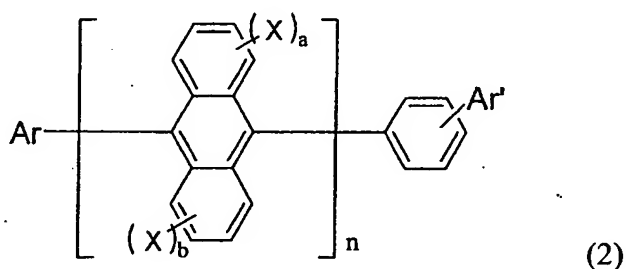
(Ar<sub>1</sub> is an (un)substituted aromatic group of 6-50 nuclear carbon atoms.)

3. The organic electroluminescence device described in Claim 1, wherein the luminescence layer contains the anthracene derivative represented by general formula (1) as the main component.

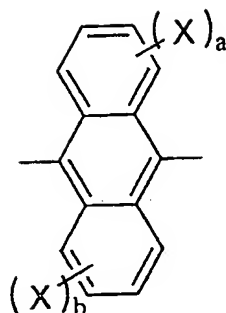
4. The organic electroluminescence device described in Claim 1, wherein the luminescence layer further contains an arylamine compound.

5. The organic electroluminescence device described in Claim 1, wherein the luminescence layer further contains a styrylamine compound.

6. Anthracene derivative represented by general formula (2) shown below:



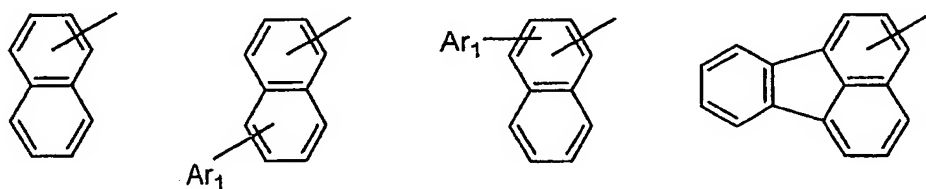
(in the formula, Ar is an (un)substituted condensed aromatic group of 10-50 nuclear carbon atoms; Ar' is an (un)substituted aromatic group of 6-50 nuclear carbon atoms; X is an (un)substituted aromatic group of 6-50 nuclear carbon atoms, (un)substituted aromatic heterocyclic group of 5-50 nuclear carbon atoms, (un)substituted alkyl group of 1-50 carbon atoms, (un)substituted alkoxy group of 1-50 carbon atoms, (un)substituted aralkyl group of 6-50 carbon atoms, (un)substituted aryloxy group of 5-50 nuclear carbon atoms, (un)substituted arylthio group of 5-50 nuclear carbon atoms, (un)substituted alkoxycarbonyl group of 1-50 carbon atoms, carboxy group, halogen atom, cyano group, nitro group, or hydroxy group; a, b, and c are whole numbers of 0-4; and n is a whole number of 1-3; when n is 2 or more, the formula inside the parenthesis shown below may be the same or different:



7. The anthracene derivative described in Claim 6, being material for an organic electroluminescence device.

8. Organic electroluminescence device having at least one luminescence-layer-containing organic thin film layer sandwiched between an anode and cathode, wherein at least one layer of the organic thin film layer contains the anthracene derivative represented by general formula (2) described in Claim 6, alone or as a component in a mixture.

9. The organic electroluminescence device described in Claim 8, wherein Ar in general formula (2) is selected from the general formulas given below:



(Ar<sub>1</sub> is an (un)substituted aromatic group of 6-50 nuclear carbon atoms.)

10. The organic electroluminescence device described in Claim 8, wherein the luminescence layer contains the anthracene derivative represented by general formula (2) as the main component.

11. The organic electroluminescence device described in Claim 8, wherein the luminescence layer further contains an arylamine compound.

12. The organic electroluminescence device described in Claim 8, wherein the luminescence layer further contains a styrylamine compound.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/10402

## A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl<sup>7</sup> C09K11/06, H05B33/14, C07C13/66, C07C15/28

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl<sup>7</sup> C09K11/06, H05B33/14, G03G5/06, C07C13/66, C07C15/28

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
CA (STN), REGISTRY (STN)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	JP 2002-329580 A (Canon Inc.), 15 November, 2002 (15.11.02), Claims; Par. No. [0083] (Family: none)	1, 3-8, 10-12
X A	WO 02/14244 A1 (Mitsui Chemicals, Inc.), 21 February, 2002 (21.02.02), & JP 2002-154993 A & EP 1221434 A1 & US 2003/0087126 A1	1, 3-8, 10-12 2, 9
X A	JP 2001-223082 A (Toray Industries, Inc.), 17 August, 2001 (17.08.01), Claims; Par. Nos. [0030] to [0036] (Family: none)	1, 3-8, 10-12 2, 9

☒ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

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"&amp;" document member of the same patent family

Date of the actual completion of the international search  
11 November, 2003 (11.11.03)Date of mailing of the international search report  
25 November, 2003 (25.11.03)Name and mailing address of the ISA/  
Japanese Patent Office

Authorized officer

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Telephone No.

Form PCT/ISA (210) (second sheet) (July 2009)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/10402

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	US 2002/0048688 A1 (Idemitsu Kosan Co., Ltd.), 25 April, 2002 (25.04.02), Claims; page 6 & WO 01/076323 A1 & JP 2001-284050 A & EP 1191822 A1	1, 3-8, 10-12 2, 9
X Y	EP 1009044 A2 (EASTMAN KODAK CO.), 14 June, 2000 (14.06.00), & JP 2000-182776 A & US 2002/0028346 A1	1, 2, 6-9 3-5, 10-12
Y	JP 11-3782 A (Toppan Printing Co., Ltd.), 06 January, 1999 (06.01.99), Claims (Family: none)	3-5, 10-12
Y	JP 10-294179 A (Mitsubishi Chemical Corp.), 04 November, 1998 (04.11.98), Claims (Family: none)	3-5, 10-12
Y	WO 02/038524 A1 (Idemitsu Kosan Co., Ltd.), 16 May, 2002 (16.05.02), Claims & JP 2001-335516 A & EP 1333018 A1	3-5, 10-12

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